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HIGH PRESSURE STUDY OF FERROMAGNETIC US*

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Resumé.--Nous avons étudié la variation avec la pression hydrostatique de la susceptibilité ac magnétique d'un échantillon polycristallin du ferromagnétique Sulfure d'Uranium (US). Sa température Curie décroît linéairement de 180,0 K, à la pression atmosphérique, jusqu'à 175,9 K à 17,6 kbar.

Abstract.--We have investigated the variation of the ac magnetic susceptibility of a polycrystalline ferromagnet US as a function of hydrostatic pressure. The Curie temperature decreases linearly from 180.0 K at the atmospheric pressure to 175.9 K at 17.6 kbar.

1. Introduction.--In the actinide series the greater extent of the 5f atomic wavefunctions implies that the crystal field interactions are greater than those in the lanthanide series. This large crystal-field potential leads to a breakdown of Russell-Saunders coupling. The difficulties in understanding the magnetism of the actinide are further enhanced by the absence of any spontaneous magnetism of the early elements (U, Np, and Pu) in the series. Nonetheless, compounds of these elements, especially those with elements of group VIA (S, Se, Te) show ferromagnetic ordering.^{1,2} All these monochalcogenides crystallize with the NaCl crystal structure. They are fairly good metallic conductors.

Guided by the unusual pressure dependence of the superconducting transition temperature of uranium metal,³ Huber, et al⁴ investigated the pressure effects on ferromagnetic UPt. They found that the saturation magnetic moment was reduced by more than 90% at 20 kbar and that the Curie temperature was pressure independent.

Our investigation on the pressure effect on ferromagnetic US was stimulated by these interesting properties of uranium compounds.

*Work performed under the auspices of the USDOE.

**Summer Research Assistant.

2. Experimental Results.--The uranium monosulfide used in this experiment was prepared by the direct reaction of uranium metal with elemental sulfur. Uranium turnings with sufficient amount of sulfur to produce a slightly hyperstoichiometric monosulfide and a small amount of iodine to boost the reaction are placed in a magnesium oxide crucible which in turn are placed in a sealed stainless steel reaction vessel. The crucible portion of the reaction vessel is placed in a pot furnace and slowly heated until the reaction starts (500-600°C). The charge is then cooled to room temperature. The reaction product can then be melted in an argon arc furnace. This second melting consolidated the reaction product and, at the same time, removed any excess sulfur in the product.

A high pressure cell with the piston-cylinder arrangement designed by C. W. Chu was employed in this study. High pressure was generated at room temperature using a one-to-one mixture of isoamyl alcohol and n-pentane to transmit the pressure. The pressure was determined by a superconducting Pb manometer. The isobaric runs were made by locking the pressure at room temperature using a Be-Cu clamp before varying the temperature of the sample and the clamp.

Fig. 1 shows the measured Curie temperatures, T_C , which we defined to be the temperatures of the maxima of the ac susceptibility. In contrast to UPt, T_C for US decreases linearly from 180.0 K at the atmospheric pressure to 175.9 K at 17.6 kbar. We have also analyzed our susceptibility data using⁵ $\chi = \text{constant} + C/(T - \theta)$, where C is the Curie constant and θ the paramagnetic Curie temperature. Our data revealed that θ decreased with pressure. The magnetic moment at 17.6 kbar decreased ~ 30% from its value at the atmospheric pressure.

3. Discussions.--First, the simplest theory for describing the actinide MCl_2 -structure compound is the one in which an integral number of f electrons on the actinide is assumed. Accordingly, Grunzweig-Genossar, et al⁶ took uranium in uranium chalcogenides to be U^{4+} with a $5f^2$ configuration and with the total an-

gular momentum of $J = 4$ in the ground state as a good quantum number. Contrastingly, because the spatial extent of 5f electrons is considerably greater than that of the 4f electrons in lanthanides and the band approach is more appropriate than the localized magnetic moment approximation, Davis⁷ has made the band calculations and his results are consistent with the photoemission work on US by Eastman and Kuznietz⁸ who concluded that the Fermi surface is located in a broad f-d band which is almost indistinguishable from the conduction band. Following Goodenough's idea⁹ of a critical interatomic distance beyond which electrons are localized and under which they are itinerant, Hill¹⁰ combined the localized and band approaches by proposing a critical value for the actinide-actinide spacing beyond which magnetic ordering results.

From the discussions above, it is clear that our data support the band picture - i.e. under high pressure, the U-U spacing in US decreases, resulting in a broadening of the f band and an increase in the f electron participation in the bonding of the metal. This "delocalization" of the f electrons gives rise to a reduction in the magnetic moment at high pressure.

Extrapolation of our data indicates in the T_c that the 5f electrons could be completely delocalized at ~ 1 megabar pressure. It is interesting to speculate whether the system might become superconducting at this point similar to Ce.¹¹

Acknowledgement. -- We would like to thank C. W. Chu for allowing us to use his high pressure cell.

Figure Caption

Fig. 1. Curie temperature, T_c , versus pressure of US.

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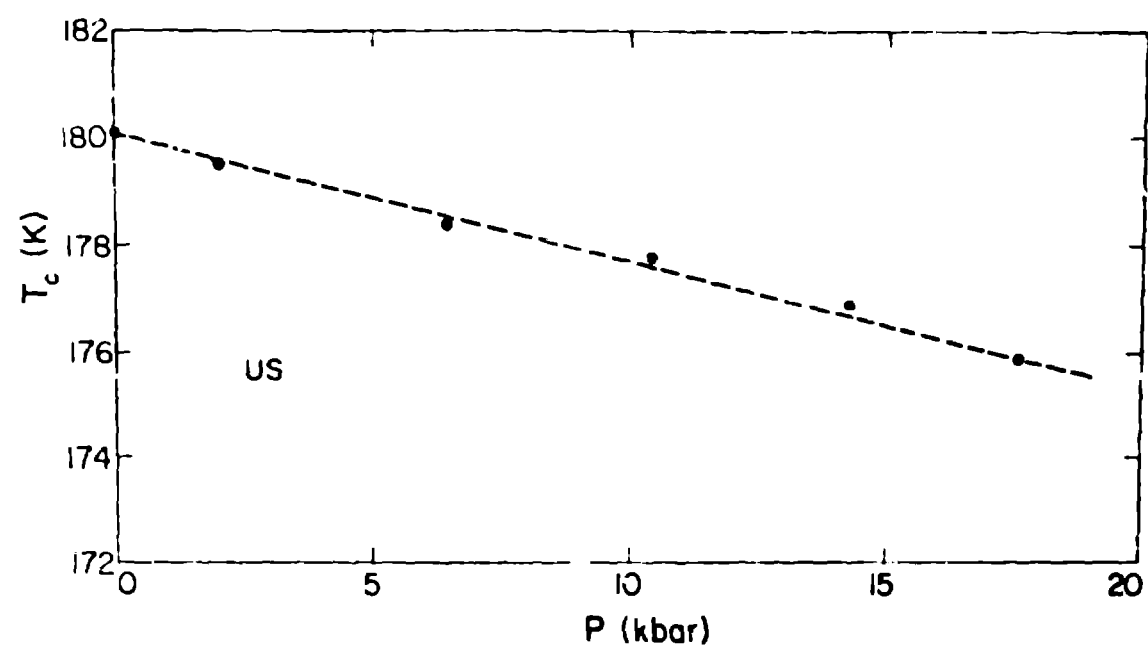


Fig. 1
Huang, Lasiewski,
Olsen, and Smith